Reprinted from Journal of the Optical Society of America, Vol. 73, page 1562, November 1983

Wavelength-dependent refractive and absorptive terms for propagation in small-spaced correlated distributions

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Received April 29, 1983

Earlier results for coherent propagation of light in correlated random distributions of dielectric particles of radius a (with minimum separation $b \geq 2a$ small compared with wavelength $\lambda = 2\pi/k$) are generalized to obtain the refractive and absorptive terms to order $(ka)^2$. The present results include the earlier multiple scattering by electric dipoles as well as scattering and multipole coupling by magnetic dipoles and electric quadrupoles. The correlation aspects are determined by the statistical-mechanics radial distribution function $f(R)$ for impenetrable particles of diameter b. The new terms for slab scatterers and spheres involve the integral of fR (first moment) or of $f \ln R$ for cylinders. The new packing factor is evaluated exactly for slabs as a simple algebraic function of the volume fraction w, and it is shown that the bulk index of refraction reduces to that of one particle in the limit $w = 1$. A similar result is achieved for spheres in terms of the Percus–Yevick approximation and the unrealizable limit $w = 1$.

INTRODUCTION

Earlier papers[1,2] developed simple forms for the coherent bulk index of refraction ($\eta = \sqrt{\epsilon}$) for correlated random distributions of dielectric particles with minimum separation (b) of centers small compared with wavelength ($\lambda = k/2\pi$). Writing the index as $\eta = \eta_r + i\eta_a + i\eta_s$, we applied general scattering theory[3,4] to the range of small kb to obtain results for the refractive (η_r) and absorptive (η_a) terms that were explicitly independent of λ and to obtain corresponding results for the scattering (η_s) loss term to lowest order in λ. The explicit approximations[2] for η_r and η_a for spheres, cylinders, and slabs ($m = 3, 2, 1$, respectively) depended only on the particles' radius or half-width (a), their complex index of refraction ($\eta' = \sqrt{\epsilon'}$), and their average number (ρ) per unit volume; they exhibited the statistical aspect of the problem only in the volume fraction $w = \rho v$, with $v = v(a)$ as the volume of one particle. The corresponding scattering terms η_s were additionally dependent on $(ka)^m$ and on the low-frequency limit of the structure factor $\mathcal{W}(W)$, with $W = \rho v(b/2) = w(b/2a)^m$ as the volume fraction of impenetrable statistical particles with diameter $b \geq 2a$, i.e., in general, each dielectric particle was visualized as having a transparent coating of thickness $(b/2) - a$. The present paper applies the general theory[3,4] to derive the leading λ-dependent terms of η_r and η_a; these depend explicitly on $(ka)^2$ and a/b for all cases and on appropriate correlation integrals $\mathcal{N}(w)$.

The correlation aspects of the distribution that we consider are determined by the statistical-mechanics radial distribution function[5] $f(R)$ for impenetrable particles and are exhibited explicitly as simple integrals over all R of the total correlation function $F = f - 1$. The integrals for spherical and slab particles are of the form $\int FR^n \mathrm{d}R$ (moments of F), but cylindrical particles also involve $\int F(\ln R)\mathrm{d}R$. These can all be evaluated numerically from existing statistical-mechanics results or approximations[5-16] for f. We obtained explicit closed-form approximations before[1,17] for the integrals that arise in the $\mathcal{W}$ set and also used the required $\mathcal{N}$ integral for spheres in a related development[18] for large kb; for slabs, we obtain both the $\mathcal{W}$ and $\mathcal{N}$ integrals from our earlier Laplace transformation[14] of the exact Zernike–Prins result[13] for f. For cylinders, we may use the virial expansion for f to consider some of the properties of $\mathcal{N}$.

In the following, for brevity, we use, for example, form (4:113) to indicate Eq. (113) of Ref. 4, as well as essentially the same notation as before.[1-4] We generalize the earlier multiple-scattering electric-dipole approximations for $\eta^2 = \epsilon$ given collectively in form (1:44) by including scattering (and multipole coupling) by magnetic dipoles and electric quadrupoles (for spheres and cylinders). For slabs and $b = 2a$ (minimum separation of slab centers equal to slab thickness), the explicit approximation for $\eta^2 = \epsilon$ reduces to $\epsilon \to \epsilon'$ if $w \to 1$, as required from physical considerations: The particles occupy all space. The limit $w \to 1$ is not realizable for identical spheres, and we take $w \lesssim w_d \approx 0.63$, with w_d as the densest random packing introduced earlier[1,17] to define the amorphous solid. However, our explicit approximation for ϵ for spheres also reduces to ϵ' as w reduces to 1; we regard this as consistent with the approximations involved in scaled-particle[6] and Percus–Yevick[5,7] statistical-mechanics theory and with the closure approximation used in the multiple-scattering theory.[3,4] For cylinders, we take $w \lesssim w_d \approx 0.84$, as before.[1] Were an analogous closed form available for the $\mathcal{N}$-integral for this case, we would expect the corresponding approximation for ϵ to show the same behavior for the nonrealizable limit $w \to 1$.

The present application of the general theory[3,4] to larger kb than before[1,2] plus the recent applications[18] to large kb provide simple forms that explicitly display the functional dependence on all key parameters for many practical applications. Thus, in these ranges of kb, elaborate machine computations are no longer required, and the results help to delineate the fundamental physical processes.

PRELIMINARY CONSIDERATIONS

For a slab-region distribution and a normally incident wave $\phi e^{-i\omega t}$ (representing either the electric or the magnetic component) we write

0030-3941/83/111562-06$01.00

$$\boldsymbol{\phi} = \hat{\mathbf{e}}e^{ikz}, \quad \hat{\mathbf{e}}\cdot\hat{\mathbf{z}} = 0, \qquad k = 2\pi/\lambda = 2\pi\eta_e/\lambda, \tag{1}$$

with η_e as the index of the embedding medium. The corresponding bulk coherent propagation coefficient

$$K = k\eta_b/\eta_e = k\eta, \quad \eta^2 = \epsilon \tag{2}$$

is to be expressed in terms of ρ and F for pair-correlated particles specified by their isolated scattering amplitudes $\mathbf{g}(\hat{\mathbf{r}}, \hat{\mathbf{z}})$. The normalization for $\mathbf{g}$ is such that for lossless particles

$$-\mathrm{Re}\,\hat{\mathbf{e}}\cdot\mathbf{g}(\hat{\mathbf{z}}, \hat{\mathbf{z}}) = -\mathrm{Re}\,g = \mathcal{M}|g(\hat{\mathbf{r}}, \hat{\mathbf{z}})|^2, \tag{3}$$

with $\mathcal{M}$ as the mean over all directions of observation $\hat{\mathbf{r}}$. The corresponding known[19] scattering coefficients a_n (which may represent two sets) are normalized by the form

$$g = \sum a_n, \quad a_n = a_n(\epsilon', x), \qquad \epsilon' = \epsilon_p/\epsilon_e, \quad x \equiv ka. \tag{4}$$

In addition to the dependence on ϵ' (the relative dielectric parameter) and on x (the normalized radius or half-width), the coefficients depend on the dimensionality (m) of the problem and on the choice of field component for $m = 2$ (i.e., on whether the electric polarization is lateral or transverse to the cylinder's axis). We obtain results for the bulk relative dielectric parameter ϵ in the form

$$\epsilon = \epsilon_1 + \epsilon_c + i\epsilon_s = \mathcal{E}_1 + x^2\mathcal{E}_c + ix^m\mathcal{E}_s, \tag{5}$$

where the set $\mathcal{E}$ is independent of $x = ka$. The forms for $\mathcal{E}_1$ and $\mathcal{E}_s$, corresponding to multiple scattering by electric dipoles, were discussed before[1,2] in detail. Now we obtain $\mathcal{E}_c$.

From Rayleigh's results for spherical dipoles,[20] the first approximation for sparse uncorrelated distributions corresponds to

$$\eta_R - 1 = -cg/2, \quad \epsilon_R = \eta_R{}^2; \quad c = \frac{i4\pi\rho}{k^3}, \quad \frac{i4\rho}{k^2}, \quad \frac{i2\rho}{k}. \tag{6}$$

In first of the papers cited in Ref. 1, for lossless small $\epsilon' = 1$, we multiplied Im η_k by the statistical-mechanics packing factor $\mathcal{W}$ to obtain the appropriate η_s for the correlated case. The complete ϵ_1 for spheres and slabs was given by Maxwell,[21] and for cylinders for both polarizations by Rayleigh.[22] We obtained ϵ_1 and ϵ_s from forms (3:74) and (4:52)

$$\eta^2 - 1 = -cG, \quad \eta^2 = \epsilon, \tag{7}$$

with G as a multiple-scattering amplitude. This form with $G = g$ was obtained originally by Reiche[23] and by Foldy[24] for spherical cases, and Lax[25] derived the form in terms of a more general amplitude than g. The function G that we require is discussed in detail in Refs. 3 and 4. In particular for spheres, cylinders, and slabs, respectively, the systems of algebraic equations (4:113), (3:92), and (3:179) determine $\epsilon = \eta^2$ functionally in terms of a_n and F for arbitrary ϵ' and $ka = x$. Before, we kept only the electric-dipole coefficient a_1 in these systems and considered only the leading terms of their imaginary and real parts, of order x^m and x^{2m}, respectively, for lossless scatterers. Now we include terms to order x^{m+2} for the refractive and absorptive effects.

Thus, for spheres with x small, the electric dipole approximates[19]

$$a_1 \approx \frac{ix^3(\epsilon' - 1)}{\epsilon' + 2}\left[1 - \frac{x^2 3(2 - \epsilon')}{5(\epsilon' + 2)}\right] - \frac{x^6 2(\epsilon' - 1)^2}{3(\epsilon' + 2)^2}, \tag{8}$$

where the next terms are of order ix^7 and x^8. The corresponding magnetic dipole ($a_{1\mathrm{M}}$) and electric quadrupole are, respectively,

$$a_{1\mathrm{M}} \approx \frac{ix^5(\epsilon' - 1)}{30}, \quad a_2 \approx \frac{ix^5(\epsilon' - 1)}{6(2\epsilon' + 3)}, \tag{9}$$

where the next terms are proportional to ix^7, ix^9, and x^{10}. The resulting amplitude for Rayleigh's approximation (6) is $g \approx a_1 + a_{1M} + a_2$. (We use a_n, a_{1M} for the earlier[4] b_n, c_1.)

For cylinders[19] and polarization transverse to the axis, we have $g \approx a_0 + a_1 + a_2$ with dominant dipole

$$a_1 \approx \frac{i\pi x^2(\epsilon' - 1)}{2(\epsilon' + 1)}\left\{1 - \frac{x^2[3 + \epsilon' - 4(\epsilon' - 1)L]}{8(\epsilon' + 1)}\right\} - \frac{x^4\pi^2(\epsilon' - 1)^2}{8(\epsilon' + 1)^2}, \tag{10}$$

where $L = \ln(2/xc')$ with $c' = 1.781\ldots$ The next terms are of order ix^6 and x^6. We also retain

$$a_0 \approx \frac{i\pi x^4(\epsilon' - 1)}{32}, \quad a_2 \approx \frac{i\pi x^4(\epsilon' - 1)}{16(\epsilon' + 1)}, \tag{11}$$

where the next terms are of order ix^6, ix^8, and x^8. For cylinders and polarization along the axis, we use $g \approx a_0 + a_1$, with

$$a_0 \approx \frac{i\pi^2(\epsilon' - 1)}{4}\left\{1 - \frac{x^2}{8}[3 - \epsilon' - (\epsilon' - 1)4L]\right\} - \frac{x^4\pi^2(\epsilon' - 1)^2}{16}, \tag{12}$$

where the next terms are of order ix^6 and x^6. In addition, we keep

$$a_1 \approx i\pi x^4(\epsilon' - 1)/16 \tag{13}$$

and ignore ix^6, ix^8, and x^8 terms.

For normal incidence on slabs, $g = a_0 + a_1$, as discussed for (3:193). The dominant term is

$$a_0 \approx ix(\epsilon' - 1)\left[1 - \frac{x^2(2\epsilon' - 1)}{3}\right] - x^2(\epsilon' - 1)^2, \tag{15}$$

where the next terms are of order ix^5 and x^4. We also retain

$$a_1 \approx ix^3(\epsilon' - 1)/3, \tag{16}$$

but not ix^5, ix^7, and x^6 terms.

DISTRIBUTION OF SLABS

From form (3:177) we have

$$G = A_0 + A_1, \quad c = i2\rho/k = iw/x, \ w = \rho 2a = \rho v(a), \tag{17}$$

where

$$A_0 = a_0(1 + A_0\mathcal{H}_0 + A_1\mathcal{H}_1/\eta),$$
$$A_1 = \eta^2 a_1(1 + A_0\mathcal{H}_1/\eta + A_1\mathcal{H}_{11}/\eta^2), \tag{18}$$

with $\mathcal{H}_{11} = c + \mathcal{H}_0$, and $\mathcal{H}_0$ and $\mathcal{H}_1$ given as functions of k, η, and F in form (3:177). This algebraic system is valid[3] for all

$ka = x$, but we consider only forms (15) and (16) for a_n and the corresponding leading terms of the correlation integrals $\mathcal{H}_n = \mathcal{J}_n + i\mathcal{N}_n$. We have

$$\mathcal{H}_0 \approx 2\rho \int_0^\infty F dR + ik2\rho \int_0^\infty FR dR \equiv \mathcal{W} - 1 + ixN,$$
$$\mathcal{H}_1 \approx -ix\eta N, \quad (19)$$

where the next terms are $0(x^2)$. Substituting into the appropriate form (7) yields

$$\epsilon = [1 - ca_0(1 + a_0\mathcal{H}_0)](1 - ca_1) \quad (20)$$

to $0(x^2)$. Thus in the corresponding form (5)

$$\epsilon = 1 + w\delta - x^2 w\delta^2[2 - w + 3N]/3 + ix\delta^2 w\mathcal{W}, \quad \delta = \epsilon' - 1, \quad (21)$$

we require only the functions $\mathcal{W}$ and N.

The rigorous pair function ρf for one-dimensional impenetrable statistical-mechanics particles of width b was given by Zernike and Prins,[13] and its Laplace transform was applied[14] in the development of a residue series. From the Laplace transform of f as in Ref. 4, we obtain the moments of $F = f - 1$ by a Taylor-series expansion:

$$F_0 = \int_0^\infty F dR = b(-2 + W)/2 \equiv -b\overline{F}_0,$$
$$W = \rho b = \rho v(b/2) = wb/2a,$$
$$F_1 = \int_0^\infty FR dR = -b^2(1 - 4W/3 + W/2)/2 \equiv -b^2\overline{F}_1, \quad (22)$$

where the notation is the same as in Ref. 18. (The identical results follow on integrating the virial expansion of F in powers of W.) Substituting into $\mathcal{W} = 1 + 2\rho F_0$ and $N = (2\rho/a)F_1$, we obtain

$$\mathcal{W} = (1 - W)^2, \quad N = -(b/a)W(1 - 4W/3 + W^2/2), \quad (23)$$

where $\mathcal{W}$ was obtained earlier[1] from the rigorous Tonks equation of state[16] (which also follows from scaled-particle theory[6]) by using statistical-mechanics theorems.

From elementary physical considerations, if $b = 2a$ (minimum separation of particle centers equal to particle width), then for $w \to 1$ (the limit of a uniform slab) we require that $\epsilon = \epsilon'$. Since $\mathcal{W} \to 0$ and $N \to -1/3$, the result (21) reduces to $\epsilon = \epsilon'$, as required.

More generally for $b = 2a$, the bracketed function in Eq. (21) reduces to

$$[\quad] = 2 - 7w + 8w^2 - 3w^3, \quad b = 2a. \quad (21')$$

DISTRIBUTION OF CYLINDERS

For polarization along the axes, to the orders of accuracy indicated for forms (12) and (13), we follow the development for slabs with

$$c = i4\rho/k^2 = i4w/\pi x^2, \qquad w = \rho\pi a^2 = \rho v(a), \quad (24)$$

where $\mathcal{H}_{11} = (c + \mathcal{H}_0 + \mathcal{H}_2)/2$ in terms of the correlation integrals in form (3:71). We have

$$\mathcal{H}_0 \approx 2\pi\rho \int FR dR + i4\rho \int F \ln(c'kR/2) R dR \equiv \mathcal{W} - 1 + i\mathcal{N},$$
$$\mathcal{H}_n \approx -i(\eta^n 2\pi\rho/n\pi) \int Fr dR = -i\eta^n(\mathcal{W} - 1)/n\pi, \quad (25)$$

where the next terms are $0(x^2)$. We obtain form (20) to $0(x^2)$, from which

$$\epsilon = 1 + w\delta + x^2 w\delta^2(1 + 2w + 4M)/8 + i\pi x^2\delta^2 w\mathcal{W}/4, \quad \delta = \epsilon' - 1. \quad (26)$$

Here

$$\mathcal{W} = 1 + 2\pi\rho \int FR dR = 1 + 2\pi\rho F_1 = 1 - 8W\overline{F}_1,$$
$$W = \pi\rho(b/2)^2 + \rho v(b/2) = w(b/2a)^2, \quad (27)$$

and

$$M = L - \pi\mathcal{N}/2$$
$$= \ln(b/a) + \mathcal{W}\ln(2/c'kb) - 2\pi\rho \int F \ln(R/b) R dR$$
$$\equiv \ln(b/a) + \mathcal{W}L_b + 8W\overline{F}_l,$$
$$\overline{F}_l = -\int_0^\infty F(\ln u) u du, \quad (28)$$

with $F(R) = F[R/b] = F[u]$ and $L_b = \ln(2/c'kb)$.

We may evaluate $\overline{F}_1$ and $\overline{F}_l$ numerically by using tabulated values of f or the original integral-equation approximations in the computing routine. To first order in W, we use the virial expansion: $F = -1$ for $u < 1$,

$$F = \frac{8W}{\pi}\left\{\cos^{-1}\frac{u}{2} - \frac{u}{2}\left[1 - \left(\frac{u}{2}\right)^2\right]^{1/2}\right\}, \qquad 1 \le u \le 2, \quad (29)$$

and $F = 0$ for $u > 2$. A closed-form approximation of $\mathcal{W}$ (and consequently of F_1) was derived earlier[1] by differentiating the scaled-particle equation of state.[6] Thus we obtained[1]

$$\mathcal{W} = (1 - W)^3/(1 + W), \quad (30)$$

from which $\mathcal{W} = 1 - 4W + 7W^2 + \ldots$. The rigorous virial expansion to $0(W^2)$ is

$$\mathcal{W} = 1 - 4W + \sqrt{3}\,12W^2/\pi \approx 1 - 4W + 6.6159W^2. \quad (30')$$

For the unrealizable value $W = 1$, the closed-form $\mathcal{W}$ vanishes; a comparable approximation of M would reduce to $-3/4$ for $b = 2a$ and $W = 1$ in order for ϵ to equal ϵ'. The corresponding moments are then $\overline{F}_1 = 1/8$ and $\overline{F}_l = -(\ln 2)/2$.

For polarization transverse to the axes we use $G = A_0 + A_1 + A_2$ with A_n satisfying the system (3:89). From the solution (3:90) and the corresponding form of η^2, in terms of a_n and $\mathcal{H}_n$ of form (3:91), to the accuracy required for present purposes, we work with

$$-(\epsilon - 1)/c = a_0 + \epsilon\mathcal{A}_1 + \epsilon^2 a_2(1 + \mathcal{A}_1 c/2)^2; \quad (31)$$
$$\mathcal{A}_1 = a_1/(1 - a_1\mathcal{H}_{11}),$$
$$2\mathcal{H}_{11} = c + (\mathcal{W} - 1) + i\mathcal{N} - i\epsilon(\mathcal{W} - 1)/2\pi,$$

where $\epsilon\mathcal{A}_1$ is a multiple-scattering coefficient that includes all electric-dipole-dipole coupling. The coefficient a_0 (essentially the magnetic dipole) is uncoupled, and the multiplier of the electric quadrupole a_2 includes all orders of electric-dipole coupling to the required accuracy.

Using approximations (10) and (11), we obtain initially

$$\epsilon_1 = 1 + \frac{2w(\epsilon' - 1)}{1 + \epsilon' - w(\epsilon' - 1)} = 1 + \frac{w\delta}{\mathcal{D}},$$
$$\mathcal{D} = 1 + \frac{(1 - w)\delta}{2}, \quad \delta = \epsilon' - 1. \quad (32)$$

with which

$$\epsilon = \epsilon_1 - \frac{x^2 w\delta}{8}\left\{\frac{3 + \epsilon' - \delta[4M + \epsilon_1(\mathcal{W} - 1)]}{2\mathcal{D}^2} - \epsilon_1 - \frac{\epsilon_1(\epsilon_1 + 1)^2}{2(\epsilon' + 1)}\right\} + \frac{i\pi x^2\delta^2 w\mathcal{W}}{8\mathcal{D}^2}. \quad (33)$$

For $b = 2a$ and the unrealizable value $w = 1$, we have $\mathcal{D} = 1$ and $\epsilon_1 = \epsilon'$, and the result for $\mathcal{W} = 0$ and $M = -3/4$ is again $\epsilon = \epsilon'$.

For comparison with form (26), say, ϵ_l, we have to $0(\delta^2)$ for the present ϵ_t,

$$\epsilon \approx 1 + w\delta - w(1 - w)\delta^2/2 + x^2 w\delta^2(1 + 2w + 4M + \mathcal{W})/16 + i\pi x^2\delta^2 w\mathcal{W}/8. \quad (34)$$

The corresponding bipolarization[2] is

$$\epsilon_l - \epsilon_t \approx w(1 - w)\delta^2/2 + x^2 w\delta^2(1 + 2w + 4M - \mathcal{W})/16 + i\pi x^2\delta^2 w\mathcal{W}/8, \quad (35)$$

and we may obtain higher-order terms in δ from Eq. (33).

DISTRIBUTION OF SPHERES

For spheres we use form (7) with

$$G = A_1 + A_{1M} + A_2, \qquad c = i4\pi\rho/k^3 = i3w/x^3,$$
$$w = \rho 4\pi a^3/3 = \rho v(a), \quad (36)$$

with the A's satisfying the system (4:113) in terms of the isolated coefficients a_n and the correlation integrals $\mathcal{H}_n$ of form (4:80) or (3:148). Introducing the low-frequency forms (3:149), we solve the system and obtain

$$-(\eta^2 - 1)/c = \eta^2\mathcal{A}_1 + \eta^2 a_{1M}[1 + \mathcal{A}_1 c\eta/(\eta + 1)]^2 + \eta^4 a_2(1 + \mathcal{A}_1 c3/5)^2;$$
$$\mathcal{A}_1 = a_1/(1 - a_1\mathcal{H}_{11}),$$
$$3\mathcal{H}_{11} = 2c + 2\mathcal{H}_0 + \mathcal{H}_2 \approx 2c + 2(\mathcal{W} - 1) + iN(2 + \eta^2/5)/x, \quad (37)$$

with

$$\mathcal{W} = 1 + 4\pi\rho\int_0^\infty FR^2\mathrm{d}R = 1 + 4\pi\rho F_2 = 1 - 24W\overline{F}_2,$$
$$W = \rho 4\pi(b/2)^3/3 = \rho v(b/2) = w(b/2a)^3, \quad (38)$$

and

$$N = -4\pi\rho a\int_0^\infty FR\mathrm{d}R = -4\pi\rho a F_1 = 24(a/b)W\overline{F}_1. \quad (39)$$

Here $\eta^2\mathcal{A}_1/a_1$ includes all electric-dipole–dipole coupling, and the multipliers of the magnetic dipole a_{1M} and the electric quadrupole a_2 incorporate multipole coupling with all orders of electric dipoles to the required accuracy.

Using approximations (12) and (13), we obtain initially

$$\epsilon_1 = 1 + \frac{3w(\epsilon' - 1)}{2 + \epsilon' - w(\epsilon' - 1)} = 1 + \frac{w\delta}{\mathcal{D}},$$
$$\mathcal{D} = 1 - \frac{(1 - w)\delta}{3}, \quad \delta = \epsilon' - 1, \quad (40)$$

with which

$$\epsilon = \epsilon_1 - x^2 w\delta\left\{\frac{1}{\mathcal{D}^2}\left[\frac{2 - \epsilon'}{5} + \frac{N}{9}\left(2 + \frac{\epsilon_1}{5}\right)\right] - \frac{\epsilon_1}{10}\left[1 + \frac{(2\epsilon_1 + 3)^2}{5(2\epsilon' + 3)}\right]\right\} + \frac{ix^3 2\delta^2 w\mathcal{W}}{9\mathcal{D}^2}. \quad (41)$$

The magnetic-dipole contribution $x^2 w\delta\epsilon_1 \propto a_{1M}\epsilon_1$ shows that the effects of the function in brackets in Eq. (37) have canceled.

By differentiating the scaled-particle approximation[6] for the equation of state, we showed that[1,17]

$$\mathcal{W} = (1 - W)^4/(1 + 2W)^2, \quad (42)$$

which also follows from the Percus–Yevick approximation.[7] The first moment F_1 obtained[8] from the Wertheim–Thiele solution of the Percus–Yevick integral equation[7] gives the closed form

$$N = \frac{2a}{b}\frac{6W}{1 + 2W}\left(1 - \frac{W}{5} + \frac{W^2}{10}\right). \quad (43)$$

Although the physically realizable range corresponds to $W \leq W_d \approx 0.63$, we see that, for $b = 2a$ and the unrealizable value $w = 1$, it follows that $\mathcal{W} = 0$ and $N = 9/5$; then $\epsilon = \epsilon'$, as was discussed for slabs and cylinders.

For comparison with forms (21), (26), and (34), we have to $0(\delta^2)$ for the present case of the sphere

$$\epsilon \approx 1 + w\delta - w(1 - w)\delta^2/3 + x^2 w\delta^2[6 + 3w - 5N]11/(15)^2 + ix^3 2\delta^2 w\mathcal{W}/9. \quad (44)$$

For $b = 2a$, the function in brackets reduces to

$$[\quad] = 3(2 - 5w + 4w^2 - w^3)/(1 + 2w). \quad (44')$$

BULK INDEX OF REFRACTION

We write forms (21), (26), (33), and (41) collectively as

$$\epsilon = \epsilon_1 + w\delta^2 P(x^2) + iw\delta^2 S(x^m), \delta = \epsilon' - 1, \quad (45)$$

where P and S, proportioned to x^2 and x^m, are obtained by inspection. The k-independent term

$$\epsilon_1 = 1 + w\delta/(1 + \delta D), \quad D = (1 - w)Q,$$
$$Q_1 = Q_{2l} = 0, \quad Q_{2t} = 1/2, \quad Q_3 = 1/3 \quad (46)$$

represents special cases of the result for ellipsoids.[2] The corresponding S for ellipsoids is also known,[2] and form (45) holds for all small dielectric particles (discounting a resonant multipole).

The corresponding bulk index of refraction may be written

$$\eta = [\epsilon_1 + w\delta^2(P + iS)]^{1/2} \approx \eta_1 + w\delta^2(P + iS)/2\eta_1, \quad (47)$$

with $\eta_1 = \sqrt{\epsilon_1}$. More generally, we write $\epsilon = \epsilon_r + i\epsilon_i$ and $\eta = \eta_r + i\eta_i$,

$$\begin{Bmatrix}\eta_r \\ \eta_i\end{Bmatrix} = \left[\frac{|\epsilon| \pm \epsilon_r}{2}\right]^{1/2}, \qquad |\epsilon| = [\epsilon_r^2 + \epsilon_i^2]^{1/2}. \quad (48)$$

In terms of $\Delta = \epsilon - 1$, we obtain form (2:3), so that part of the earlier development is appropriate.

In particular, if we retain ϵ_1 to $0(\delta^3)$ and P and S to $0(\delta^0)$, then

$$(\epsilon - 1)/w = \Delta/w = \delta(1 - \delta D + \delta^2 D^2) + \delta^2(P + iS) \quad (49)$$

is correct to $0(\delta^3)$, $0(x^2\delta^2)$, and $i0(x^m\delta^2)$. For complex $\delta = \delta_r + i\delta_i$ we construct $\Delta = \Delta_r + i\Delta_i$ essentially as for form (2:25) with the earlier iS replaced by $iS + P$ but retain only the leading term of the earlier δ^3 contribution. Thus

$$\Delta_r/w = \delta_r(1 - \delta_r D + \delta_r^2 D^2) + \delta_i^2 D(1 - 3\delta_r D) - 2\delta_r\delta_i S + (\delta_r^2 - \delta_i^2)P \quad (50)$$

and

$$\Delta_i/w = \delta_i[1 - 2\delta_r D + (3\delta_r^2 - \delta_i^2)D^2] + (\delta_r^2 - \delta_i^2)S + 2\delta_r\delta_i P. \quad (51)$$

The terms in P account for the $0(k^2)$ corrections indicated for forms (2.22) ff. If $\delta_i = 0$, then Δ_r is independent of S, and Δ_i of P.

Similarly in terms of $\nu = \eta' - 1$ with $\eta' = \eta_p/\eta_e$, we write $\delta = \eta'^2 - 1 = \nu(2 + \nu)$ and express $\eta - 1$ to $0(\nu^3)$, $0(x^2\nu^2)$, and $i0(x^m\delta^2)$ as

$$(\eta - 1)/w = \nu + \nu^2(A + 4P + i4S)/2 + \nu^3 B/2,$$
$$A = 1 - (w + 4D), \quad B = -(1 - w)(w + 4D) + 8D^2. \quad (52)$$

For complex $\nu = \nu_r + i\nu_i$ we construct $\eta = \eta_r + i\eta_i$ essentially as for form (2:28). Thus the refractive contrast $\eta_r - 1$ is given by

$$(\eta_r - 1)/w = \nu_r(1 + \nu_r A/2 + \nu_r^2 B/2) - \nu_i^2(A + 3\nu_r B)/2 - 4\nu_i\nu_r S' + 2(\nu_r^2 - \nu_i^2)P, \quad (53)$$

and the net attenuation is determined by

$$\eta_i/w = \nu_i[1 + \nu_r A + (3\nu_r^2 - \nu_i^2)B/2] + 2(\nu_r^2 - \nu_i^2)S + 4\nu_i\nu_r P. \quad (54)$$

If the scatterers are lossless ($\nu_i = 0$), then η_r does not depend on S, and η_i is independent of P. The present forms (50)–(54) hold for all small-spaced particles to the indicated accuracy. Generalization of D and S to aligned ellipsoidal particles and cocentered nonsimilar ellipsoidal exclusion regions are given in Ref. 2.

For measurements in which the parameters ϵ_0 and η_0 of the embedding medium are varied, we normalize ϵ_b and η_b with respect to ϵ_p and η_p instead of ϵ_0 and η_0. See Ref. 2 for details.

The values of D, P, S, A, and B for forms (49)–(54) are given by the following for the special cases at hand. For slabs,

$$D_1 = 0, \quad A_1 = 1 - w, \quad B_1 = -w(1 - w), \quad (55)$$

$$P_1 = -x^2(2 - W + 3N)/3, \quad S_1 = x\mathcal{W}, \quad (56)$$

with $\mathcal{W}$ and N as in forms (19) and (23). For cylinders and lateral polarization, $\{D_{2l}, A_{2l}, B_{2l}\}$ equals $\{D_1, A_1, B_1\}$, and

$$P_{2l} = x^2(1 + 2w + 4M)/8, \quad S_{2l} = x^2\pi\mathcal{W}/4. \quad (57)$$

For cylinders and transverse polarization,

$$D_{2t} = (1 - w)/2, \quad A_{2t} = -(1 - w) = -A_{2l}, \quad B_{2t} = -w(1 - w) = B_{2l}, \quad (58)$$

$$P_{2t} = x^2(1 + 2w + 4M + \mathcal{W})/16 = P_{2l}/2 + x^2\mathcal{W}/16, \quad S_{2t} = \pi x^2\mathcal{W}/8 = S_{2l}/2, \quad (59)$$

with M and $\mathcal{W}$ as in forms (25)–(30). For spheres,

$$D_3 = (1 - w)/3, \quad A_3 = -(1 - w)/3, \quad B_3 = -(1 - w)(4 + 5w)/9, \quad (60)$$

$$P_3 = x^2(6 + 3w - 5N)11/(15)^2, \quad S_3 = 2x^3\mathcal{W}/9, \quad (61)$$

with N and $\mathcal{W}$ as in forms (38), (39), (42), and (43).

For cylinders, the values of ϵ' and η' for the lateral and transverse cases may differ, and the corresponding birefrengence $(\eta_{li} - \eta_{ti})$ will then display intrinsic as well as form effects. See Ref. 2 for details. The relations $A_{2t} = -A_{2l}$ and $B_{2t} = B_{2l}$ simplify considerations. In particular, if there is a common ν, then

$$\eta_l - \eta_t = w\nu^2[A_l + P_l - x^2\mathcal{W}/16 + iS_l]$$
$$= w\nu^2[1 - w + x^2(1 + 2w + M - \mathcal{W})/16 + i\pi x^2\mathcal{W}/4] \equiv \nu^2(R + iI). \quad (62)$$

Then the birefrengence corresponds to

$$\mathrm{Re}(\eta_l - \eta_t) = (\nu_r^2 - \nu_i^2)R - 2\nu_r\nu_i I \quad (63)$$

and the dichroism to

$$\mathrm{Im}(\eta_l - \eta_t) = 2\nu_r\nu_i R + (\nu_r^2 - \nu_i^2)I. \quad (64)$$

For experiments in which η_e is varied, we introduce the variable $\xi = (\eta_e - \eta_{pr})/\eta_{pr}$ and the constant $\mu = \eta_{pi}/\eta_{pr}$ to construct $\nu = (-\xi + i\mu)/(1 + \xi)$. For small ξ, we have

$$\mathrm{Re}(\eta_{bl} - \eta_{bt})/\eta_{pr} \approx [(\xi^2 - \mu^2)R + 2\xi\mu I](1 - \xi), \quad (65)$$

$$\mathrm{Im}(\eta_{bl} - \eta_{bt})/\eta_{pr} \approx [-2\xi\mu R + (\xi^2 - \mu^2)I](1 - \xi), \quad (66)$$

which generalize the result in form (1:67).

ACKNOWLEDGMENT

This research was supported in part by grants from the National Science Foundation and the U.S. Office of Naval Research.

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